High responsivity solar blind photodetector based on high Mg content MgZnO film grown via
pulsed metal organic chemical vapor deposition

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Abstract

Metal-semiconductor-metal (MSM) structured solar blind photodetectors were fabricated based on various Mg content wurtzite $\text{Mg}_x\text{Zn}_{1-x}\text{O}$ epitaxial films grown via pulsed metal organic chemical vapor deposition. The response spectra of the devices showed a peak position that shifts from ~ 383 nm to 276 nm for Mg content, x , between 0.0 and 0.51, covering a wide portion of the ultra-violet spectral region, extending well into the solar blind window. At 10 V bias, a large responsivity of $\sim 1.8 \times 10^4$ A/W was obtained at 276 nm for a device based on a high Mg content ($x=0.51$) MgZnO film. To the best of our knowledge, this responsivity is the highest ever reported for a MgZnO based device and its origin is attributed to large internal gain resulting from carrier trapping at the MgZnO/Ni/Au interface. This is confirmed by the presence of an asymmetric Schottky barrier height on the two MSM contacts. Conversely, the response speed of the devices was slow with the 10%-90% rise and fall times measured to be in the millisecond range. The results reported in this work show the realization of high responsivity MgZnO based solar blind photodetectors, providing a significant step in the development of MgZnO alloy based of detector.

Keywords: Solar blind photodetector, MgZnO films, Responsivity, Temporal response, cutoff wavelength, Pulsed metal organic chemical vapor deposition

1. Introduction

Solar blind photodetectors with long wavelength cutoffs of ~ 290 nm are of great interest for a variety of applications such as flame sensing, missile detection, air and water purification, and medical diagnostics as well as space-to-space communications [1-6]. For such applications, wide band gap (WBG) semiconductors offer the advantages of compact size and low cost, high breakdown field, and filter free ultra-violet (UV) detection [3, 6]. Among the many WBG semiconductors are the nitrides which have been extensively studied for UV photodetector applications [1, 2, 7], but recently ZnO-MgO alloys have received great attention due to their high radiation resistance, wide operational temperature, and widely tunable band gap [8-10].

$\text{Mg}_x\text{Zn}_{1-x}\text{O}$ ($0 < x < 1$) alloys with tunable band gaps between 3.24 eV ($x=0$, wurtzite ZnO) and 7.8 eV ($x=1$, cubic MgO) are a promising material for UV photodetectors covering the entire solar blind spectral window [10, 11]. However, the limited solubility of MgO into ZnO and the structural dissimilarity between the two lead to the unavoidable problem of phase segregation, making it difficult to tune the cutoff wavelength into the deep UV spectral window [12-14]. Nevertheless, researchers have continuously been improving epitaxial film growth techniques, including pulsed laser deposition (PLD), [10, 12, 15] molecular beam epitaxy (MBE) [16-18] and metal organic chemical vapor deposition (MOCVD) [19, 20] to achieve successful growth of wurtzite MgZnO with a Mg content of up to 0.37 (4.28 eV) and cubic MgZnO with a Mg content exceeding 0.62 (5.40 eV). Alloys with a Mg composition between 0.37 and 0.62 have been found to be mixed phase material without a defined band gap thereby leading to a discontinuity in the band gap tailoring between 4.28 eV and 5.40 eV [12, 21]. This missing band

adversely affects possible UV detection applications as it covers a large portion of the solar blind spectral region.

Recently, the authors have reported on the growth of a record high Mg content (51 %) wurtzite phase MgZnO epitaxial film with a band gap of ~ 4.50 eV (276 nm) which is well within the solar blind spectral window [22, 23]. This was achieved via the pulsed metal organic chemical vapor deposition (PMOCVD) method in which fast periodic transitions between MgZnO layer growth and oxidation create a far from equilibrium process, resulting in high Mg content wurtzite MgZnO without phase segregation [24, 25]. In this paper, we report on the photodetector properties of PMOCVD grown MgZnO films with various Mg content. The films were fabricated into metal-semiconductor-metal (MSM) interdigital structured photodetector by standard liftoff processes. Results from spectral measurements demonstrate the possibility of covering a large portion of the solar blind spectral window by varying the Mg content in the film. Absolute responsivity as high as $\sim 18,000$ A/W at a peak wavelength of 276 nm was obtained for a film with the highest Mg content (51 %) at a 10 V bias. This high responsivity for MgZnO alloys is rare, to the best of our knowledge, and suggests the presence of high internal gain in the device. Comparison of the I-V characteristics under “dark” and “light” also show that the materials are photoresponsive.

2. Experimental Procedures

Mg_xZn_{1-x}O films (450 nm-890 nm) with Mg content between $x=0.0$ and $x=0.51$ were grown on ZnO (~ 30 nm)/AlN (~ 25 nm)/Al₂O₃ (0001) substrates via the PMOCVD method. Details of the experimental procedure for the growth of the MgZnO films can be found elsewhere [22, 24]. The MgZnO films were fabricated into metal-semiconductor-metal (MSM) interdigitated photodetectors. Figure 1 (top) shows a schematic of an MSM photodetector with interdigital

finger patterns 500 μm long, 10 μm wide, and 10 μm inter-electrode spacing patterned using the standard liftoff process with a Ni/Au bilayer thickness of 20 nm/130 nm. An edge view of the film is also presented in Figure 1 (bottom), showing the stacked layers of the film and substrate.

Figure 1. Schematics of MSM interdigitated structure for MgZnO photodetectors (top) with finger width and inter-electrode spacing of 10 μm , and an edge view of MgZnO films grown on ZnO/AlN/Al₂O₃ (0001) substrate (bottom).

The response spectra of the devices were measured using a 150 W Xenon lamp as an excitation source. The radiation passes through a scanning monochromator (Newport Cornerstone 130 1/8 m) and is focused onto the detector placed on a probe station. The photocurrent generated by the detector was measured by a lock-in amplifier (SR810) in sync with an optical chopper (Cornerstone 130 integrated chopper) at a frequency of 380 kHz and the devices were externally biased with a Keithly 2400 source meter. The measured spectra were then scaled by the calibrated responsivity profile and power spectrum obtained from scans performed on a 818-UV Si photodetector. The I-V characteristics under “dark” and “light” conditions were measured by the Keithly 2400 source meter contacted to the devices through

probes. The dark measurement was conducted in a darkened room, while the “light” measurements took place under illumination from the 150W Xenon lamp, at a wavelength corresponding to the band gap energy of the film (Mg content dependent) using the 1/8m monochromator. The data was collected by GPIB interface data collection software. The temporal response of the photodetectors was measured by using a deep UV LED (UVTOP270, SETI) that excites the devices at an emission wavelength of 270 nm, at or below the peak responsivity wavelength of all devices. The rise and fall time of the UV LED was <10 ns. The MSM devices were connected in series with a load resistance of 464 Ω and biased at 6 V.

3. Results and discussion

Figure 2 (a) shows the normalized response spectra of $\text{Mg}_x\text{Zn}_{1-x}\text{O}$ photodetectors with the Mg content varying from $x = 0.0$ to 0.51. Examining the spectrum, the peak position for the responsivity of the detectors shows a clear blue shift from ~383 nm ($x=0.0$) to 276 nm ($x=0.51$), extending well into the solar blind spectral window (long wavelength cutoff < 290 nm). The decrease in the peak responsivity wavelength (λ_p) with the Mg content in the film is shown in Figure 2 (b, red) and is in agreement with the blue shift in the absorption edge observed for these films [22].

Interestingly, it is clear from the spectra presented in Figure 2 (a) that a photo-response at photon energies lower than the bandgap of the material is observed for each detector. This indicates the presence of carrier transitions (sub-band response) which may result from defect levels or band tails introduced due to stacking faults during the synthesis process [26, 27]. In addition, for the alloyed films the sub-band response could result from microscopic Mg composition fluctuations in the films [11, 28].

The -3 dB cutoff wavelength ($-3\text{ dB-}\lambda_C$) above and below the peak responsivity wavelength (λ_P) for the devices is shown in Figure 2 (b) (black for $-3\text{ dB-}\lambda_C (> \lambda_P)$ and blue for $-3\text{ dB-}\lambda_C (< \lambda_P)$). Similar to λ_P , the $-3\text{ dB-}\lambda_C$ decreases with the Mg content in the film. The UV/visible rejection ratio ($R(\lambda_P)/R(400\text{ nm})$) for the devices is more than two orders of magnitude at a 10 V bias, demonstrating that the devices can potentially operate in the UV spectral window without significant interference from any background visible radiation [12, 29]. For wavelengths shorter than λ_P (high photon energies), the responsivity of the devices decreases sharply. This is attributed to the generation of hot carriers (thermalization) in which the generated electron-hole pairs thermalize within their respective bands by releasing energy via phonons, and strong absorption near the surface of the materials. When light is absorbed near the surface, it is highly likely that the recombination lifetime is very short due to surface states or defects, so that the photocarriers recombine before being collected, leading to a decrease in the responsivity of the devices [30].

Figure 2. (a) Response spectra of MgZnO UV photodetectors with various Mg content, and (b) dependence of responsivity peak position, λ_p (red), -3dB cutoff wavelength ($\lambda_c < \lambda_p$, blue), and -3dB cutoff wavelength ($\lambda_c > \lambda_p$, black) on the Mg content of the film.

The current voltage (I - V) characteristics of the devices were measured with and without illumination. Figure 3 shows typical dark (black circle) and illuminated (red circle) I - V curves for a $\text{Mg}_x\text{Zn}_{1-x}\text{O}$ ($x=0.51$) MSM photodetector. As a result of illumination, the current through the device increases, indicating the generation of higher carrier concentration in the device (i.e. the material is photoresponsive). The inset in Figure 3 shows the dependence of resistance, calculated at a 10 V bias, on Mg content in the MgZnO films from which the MSM detectors were fabricated. The resistance measured from both the dark and illuminated I - V curves showed an increase with the Mg content in the film, which is expected due to an increase in the bandgap of the material. After illumination at a wavelength $\sim \lambda_p$, the resistance generally reduced without losing its upward trend with Mg content. The non-linear behavior observed in the I - V characteristics of the detectors indicates the formation of a Schottky barrier at the MgZnO and Ni/Au bilayer junctions [26].

Figure 3. I-V characteristics of $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ photodetector under dark and illumination. The inset shows resistance calculated at a 10 V bias versus Mg content in the films. The devices with Mg content of $x=0.12$, 0.27 , 0.46 , and $x=0.51$ were illuminated at wavelengths of 350 nm, 320 nm, 290 nm, and 280 nm, respectively.

As clearly indicated in Figure 2 (a), the MSM photodetector fabricated from the $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ film had a responsivity peak within the solar blind spectral window, suggesting its potential use for various application [4, 5]. The absolute responsivity of the device measured at 5 V, 8 V, 10 V bias is shown in Figure 4 (a). At a 10 V bias, the peak responsivity ($\lambda_p=276$ nm) of the device is $\sim 18,000$ A/W. This value is comparable to that reported for pure ZnO (26 000 A/W at an 8 V bias, and 5 μm inter-electrode spacing) [9], and is higher by more than one order of magnitude than the largest value reported for MgZnO-based deep UV photodetectors (1200 A/W at 5V bias and 5 μm inter-electrode spacing) [11]. Above the -3 dB cutoff wavelength ($\lambda_c = 290$ nm), the responsivity curve shows an oscillatory shoulder whose intensity increases with an

applied bias. The origin of this shoulder is attributed to the ZnO buffer layer grown to improve the crystal quality of the film.

Figure 4. (a) Responsivity for Mg_{0.51}Zn_{0.49}O based photodetector at 5.0 V, 8.0 V, and 10.0 V bias. The inset shows the transmission spectrum of the Mg_{0.51}Zn_{0.49}O film showing a drop in transmission ~380 nm due to ZnO buffer layer. (b) Responsivity and internal gain versus bias at $\lambda_p=276$ nm.

The effect of a ZnO buffer layer was also observed in the transmission spectrum of the Mg_{0.51}Zn_{0.49}O thin film (Figure 4 (a), inset), exhibiting a sharp drop in transmittance ~380 nm [22]. To explain this further, consider the behavior of incident photon flux which falls off exponentially ($\sim \exp(-\alpha(\lambda)z)$ in a given material, where $\alpha(\lambda)$ is the absorption coefficient of the material and z is the depth [30]). The depth (penetration depth, δ) at which the photon flux in the material reduces to 37 % of its value at the surface is expressed as: $\delta=1/\alpha(\lambda)$. Calculating $\alpha(\lambda)$ from the transmission spectrum (Figure 4 (a, inset)) using the total thickness of the film of ~ 500 nm out of which ~ 30 nm is the ZnO buffer layer, one can obtain $\delta(\lambda)$. Accordingly,

penetration depths of 8.0 μm , 1.3 μm , and 0.2 μm were estimated for wavelengths of 380 nm, 300 nm, and 276 nm, respectively. This clearly indicates that for $\lambda > 300$ nm, the $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ film is transparent and the photon flux reaches the depth at which the ZnO buffer layer is located (~ 470 nm from the surface of the film) without significant reduction, leading to the observed shoulder.

As discussed above, although the ZnO buffer layer is crucial for the growth of high crystalline wurtzite MgZnO films, its narrow absorption band leads to a reduced out of band rejection ratio, degrading the performance of the device. This effect can be eliminated by replacing ZnO with a monoclinic gallium oxide ($\beta\text{-Ga}_2\text{O}_3$) buffer layer that has a higher band gap (~ 4.9 eV) [31] than the $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ film (~ 4.50 eV). Additionally, its excellent epitaxial relationship with wurtzite structure material such as GaN makes it essential for the growth of MgZnO alloys with structural quality similar to those obtained using ZnO buffer layers [32].

The dependence of peak responsivity on bias voltage is shown in Figure 4 (b, left panel). With an increase in bias, the responsivity of the device increases, indicating no carrier mobility or sweep-out effect up to the 10 V applied bias [11]. The responsivity at a given bias (R_V) of a photodetector can be expressed as [30]:

$$R_V = \frac{q\lambda}{hc} \eta G, \quad (1)$$

where η is the quantum efficiency, q is the fundamental electric charge, λ is the incident light wavelength, h is the Plank's constant, c is the speed of light, and G is an internal gain of the device. Assuming a quantum efficiency of one, which can be attained for a thick, high quality film with antireflection coating, the internal gain of the device can be estimated [9, 30]. Figure 4 (b, right panel) shows the dependence of internal gain (G) on the bias voltage and follows a trend similar to that for responsivity. Corresponding to the peak responsivity at a 10 V bias, an internal

gain of $\sim 1.3 \times 10^5$ was obtained. Such a high internal gain has been reported for pure ZnO photodetectors by Liu et al. [9], and was attributed to carrier trapping at the electrode/metal interfaces of the MSM devices. It is noteworthy that the estimated internal gain in this work is the minimum since the quantum efficiency is assumed to be one; for lower quantum efficiency the internal gain is higher than what is presented here.

The high internal gain in the MgZnO based devices could be due to carrier trapping in the film and film/metal interfaces [9, 11]. To study this, we analyzed the dark I-V characteristics of the device as shown in Figure 5. The I-V characteristics clearly shows that the carrier transport mechanism in the device is interface limited which can be described either by Schottky thermionic emission or a tunneling model [33]. Which of the two mechanisms is dominant can be determined by using a rough criterion that compares the characteristic tunneling energy (E_{00}) and the thermal energy ($k_B T$), where $E_{00} = 0.5 q \hbar \sqrt{N/m^* \epsilon_s \epsilon_0}$, k_B is the Boltzmann constant, and T is the absolute temperature. If $E_{00} < k_B T$, the Schottky thermionic emission dominates the conduction mechanisms prevailing over the tunneling process [33]. In the definition of E_{00} , q , $\hbar$, N , m^* , ϵ_s , and ϵ_0 represent electric charge, Planks constant, carrier concentration, effective mass, dielectric constant of the material, and permittivity of free space, respectively. Using $\epsilon_s = 12$, $m^* = 0.24 m_0$ reported for ZnO [34], and $N \sim 10^{16} \text{ cm}^{-3}$ measured for ZnO film by Hall measurements we obtain E_{00} of $\sim 2.0 \text{ meV} \ll k_B T$ (26 meV), suggesting that the dominant conduction mechanism at the MgZnO/Ni/Au interface is Schottky thermionic emission.

Figure 5. Dark I-V characteristics of the $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ MSM photodetector. The experimental data (open circle) is fitted to the back-to-back Schottky thermionic emission model.

Since the MSM photodetector has two Schottky barriers connected back-to-back, a bias of either polarity will put one Schottky barrier in a reverse direction and the second one in a forward direction. Therefore, the equation used to describe the current dependence on the bias voltage can be written as [33]:

$$I(V) = AA^*T^2 \left[\left(\exp \left(\frac{qV}{nk_B T} \right) - 1 \right) \exp \left(\frac{-q\phi_{b1}}{k_B T} \right) + \left(\exp \left(-\frac{qV}{nk_B T} \right) - 1 \right) \exp \left(\frac{-q\phi_{b2}}{k_B T} \right) \right]. \quad (2)$$

Here, A is the area of the MSM photodetector $\sim 2.5 \times 10^{-3} \text{ cm}^2$, A^* is effective Richardson constant which is $28.4 \text{ A cm}^{-2} \text{ K}^{-2}$ for ZnO, $q\phi_{b1}$ and $q\phi_{b2}$ are Schottky barrier heights at the two interfaces, V is the bias voltage, and n is the ideality factor. By fitting the measured I-V data (Figure 5) to Eq. (2) one can extract the Schottky barrier height at the two metal/film interfaces of the MSM photodetector. Consequently, $q\phi_{b1}$ and $q\phi_{b2}$ of 0.363 eV and 0.367 eV for the positive bias region and 0.475 eV and 0.448 eV for the negative bias region were extracted, showing an asymmetry in the Schottky barrier height at the two MgZnO Ni/Au interfaces.

According to Liu et al [9], such asymmetry in barrier height originates from the trapping of holes at the film/metal interface. The internal gain of a photodetector is defined as the ratio of the lifetime of excess carriers (τ), to the transit time (t_{tr}), (i.e $G = \tau/t_{tr}$). In the case of hole trapping at the MgZnO/Ni/Au interface electrons circulate through the external circuit multiple times before recombining with holes (i.e. $\tau \gg t_{tr}$), leading to an enhanced internal gain and, thus, high photodetector responsivity.

Figure 6. Temporal response of a $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ photodetector excited by a high power LED source with excitation wavelength of 270 nm and 50 ms pulse width. The rise and fall times for the UV LED are <10 ns.

The response speed of the MgZnO photodetectors were evaluated by measuring the temporal response of the detectors at a 6 V bias using a 464 Ω load. Figure 6 shows a representative temporal response for a $\text{Mg}_{0.51}\text{Zn}_{0.49}\text{O}$ photodetector. The 10 %-90 % rise and fall times for devices with $x = 0.27, 0.46$, and 0.51 are presented in Table I. Generally, the devices were found to be slow with both the rise and fall times to be in the millisecond range. Compared

to devices with similar MgZnO composition, our devices are ~3-10 times slower which isn't unanticipated given the high responsivity values [4, 35]. The response speed of the devices decreases with an increase in Mg content in the film which could be due to an increase in trap concentration. For these devices, the RC time limit on the response speed is negligible since the interdigital capacitance is estimated to be < 1pF. Similarly, the transit time, $t_{tr} = \frac{g^2}{V\mu_e}$, where μ_e is the electron mobility (61 cm²/Vs[22]), V is the bias voltage (6.0 V) and g is the gap between two electrode fingers is ~2.7 ns, showing no influence both on the rise and fall times of the devices. A possible factor that affects the response speed of these devices could be an excessive lifetime of trapped carriers, which in our case is likely due to the trapped holes at the MgZnO thin film and electrode interfaces as discussed above (Figure 5). When an excessive lifetime of trapped carrier affects the response speed of a device, large internal gains and high responsivity are observed [4], which is exactly the case here (see Figure 4). It is believed that the origin of trap states at the interface may be related to surface damage originating during device processing or interface states associated with microscopic fluctuation of Mg composition in the films [28]. Annealing the films before and after device fabrication may be used to minimize the trap states, and hence improve the response speed.

Table I. Rise and fall time for MgZnO photodetectors with various Mg content

Detectors	λ_{cutoff} (nm)	Rise time (ms)	Fall time (ms)
Mg _{0.27} Zn _{0.73} O	320	1.65	5.71
Mg _{0.46} Zn _{0.54} O	290	2.99	7.01
Mg _{0.51} Zn _{0.49} O	276	5.70	35.3

4. Conclusions

In conclusion, metal-semiconductor-metal (MSM) photodetectors were fabricated on wurtzite MgZnO films with various Mg content grown by pulsed metal organic chemical vapor deposition. The peak response of the devices shifted from ~383 nm ($x=0.0$) to 276 nm ($x=0.51$), extending well into the solar blind spectral window. The I-V characteristics under illumination confirmed the photoresponsive properties of the devices. Devices based on high Mg content ($x=0.51$) films had a responsivity of ~18 000 A/W at a bias of 10 V. This is the highest responsivity ever reported for a MgZnO based device and attributed to a high internal gain due to carrier trapping states at the MgZnO/Ni/Au interface, as suggested by the asymmetric barrier height at the two sides of the MSM electrodes. Temporal response measurements showed that the devices are generally slow, with 10%-90% rise and fall times in the millisecond range. The results demonstrate the realization of high responsivity solar blind photodetectors based on high Mg content MgZnO films. Such a high responsivity photodetector can be potentially used for detecting weak UV emitting sources in the solar blind window such as missile plume and flame without amplifying electronics.

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